

International Global Atmospheric Chemistry (IGAC) Project's First Aerosol Characterization Experiment (ACE 1): Overview

Timothy S. Bates,¹ Barry J. Huebert,² John L. Gras,³ F. Brian Griffiths,⁴
and Philip A. Durkee⁵

Abstract. The southern hemisphere marine Aerosol Characterization Experiment (ACE 1) was the first of a series of experiments that will quantify the chemical and physical processes controlling the evolution and properties of the atmospheric aerosol relevant to radiative forcing and climate. The goals of this series of process studies are to reduce the overall uncertainty in the calculation of climate forcing by aerosols and to understand the multiphase atmospheric chemical system sufficiently to be able to provide a prognostic analysis of future radiative forcing and climate response. ACE 1, which was conducted from November 15 to December 14, 1995, over the southwest Pacific Ocean, south of Australia, quantified the chemical, physical, radiative, and cloud nucleating properties and furthered our understanding of the processes controlling the aerosol properties in this minimally polluted marine atmosphere. The experiment involved the efforts of scientists from 45 research institutes in 11 countries.

1. Introduction

Atmospheric aerosol particles affect the Earth's radiative balance both directly by scattering and absorbing solar radiation and indirectly by modifying the optical properties and lifetime of clouds. The natural aerosol derived from biogenic sulfur and organic species, sea salt, and mineral aerosol has been substantially perturbed by anthropogenic aerosols, particularly sulfates from SO₂ emissions and organic condensates and soot from biomass and fossil fuel combustion [*Intergovernmental Panel on Climate Change (IPCC)*, 1996]. The global mean radiative forcing due to the direct effect of anthropogenic sulfate aerosol particles alone is calculated to be of comparable magnitude (approximately -0.2 to -0.8 W m⁻²) but opposite in sign to the forcing

due to anthropogenic CO₂ and the other greenhouse gases [Charlson *et al.*, 1991, 1992; Kiehl and Briegleb, 1993; Penner *et al.*, 1994; Haywood and Shine, 1995; IPCC, 1996; Chuang *et al.*, 1997]. More uncertain is the radiative forcing due to the indirect cloud-mediated effects of sulfate aerosol particles [Boucher and Lohmann, 1995]. Furthermore, the global distribution of aerosol particles is extremely inhomogeneous due to their relatively short lifetimes (in the range of 4–5 days [IPCC, 1996]). This results in a negative forcing that is focused in particular regions and subcontinental areas. This uneven forcing can cause continental to hemispheric scale effects on climate patterns.

Although aerosol particles have a potential climatic importance over and down wind of industrial regions that is equal to that of anthropogenic greenhouse gases, they are still poorly characterized in global climate models. This is a result of a lack of both globally distributed data and a clear understanding of the processes linking gaseous precursor emissions, atmospheric aerosol properties, and the spectra of aerosol optical depth and cloud reflectivity. At this time, tropospheric aerosols pose one of the largest uncertainties in model calculations of climate forcing. This uncertainty significantly limits our ability to assess the effect of natural and human-induced changes in the chemistry of the atmosphere on global climate.

¹Pacific Marine Environmental Laboratory (PMEL), NOAA, Seattle, Washington.

²Department of Oceanography, University of Hawaii, Honolulu.

³Division of Atmospheric Research, Commonwealth Scientific and Industrial Research Organisation, Mordialloc, Victoria, Australia.

⁴Division of Marine Research, Commonwealth Scientific and Industrial Research Organisation, Hobart, Tasmania, Australia.

⁵Department of Meteorology, Naval Postgraduate School, Monterey, California.

2. ACE Goals and Objectives

The overall goals of the Aerosol Characterization Experiments (ACE) are to reduce the overall uncer-

tainty in the calculation of climate forcing by aerosols and to understand the multiphase atmospheric chemical system sufficiently to be able to provide a prognostic analysis of future radiative forcing and climate response. Achieving these goals requires further development of chemical transport models to produce accurate global aerosol distributions, clear and cloudy sky radiative transfer models to calculate the radiative effects of aerosols, and climate system models to study the interaction of aerosol particles within the integrated climate system (Figure 1). Further developing and testing of these models require simultaneous measurements of aerosol chemical, physical, radiative, and cloud nucleating properties and the processes controlling those properties. Radiative transfer models require values for aerosol optical properties such as the light scattering efficiency per unit mass (α_{sp}), the upward scattered fraction ($\bar{\beta}$) or asymmetry factor (g), the fraction of light scattered versus that absorbed or single scattering albedo (ω_o) and the dependence of scattering by the aerosol on relative humidity ($f_{sp}(RH)$). All these properties depend in turn on the chemical composition, size distribution, morphology, and state of the mixture of the aerosol. Measurements of aerosol chemical and physical properties, such as the mass distribution of all chemical species, the degree of mixing of various chemical species, and the overall size distribution, are thus needed to link global aerosol distributions with aerosol optical properties. Improving chemical transport mod-

els requires a quantitative understanding of precursor gas and aerosol emissions and the processes controlling aerosol formation, transport, chemical transformation, and removal.

Reducing the uncertainties in estimates of the radiative forcing of climate by aerosols will require a combination of laboratory experiments, long-term continuous and short-term intensive field studies, satellite observations and modeling analyses [Penner *et al.*, 1994]. Although much of this work can be accomplished by single laboratory groups, intensive process studies that seek to obtain closure (internal consistency) between various measured and modeled aerosol properties [Quinn *et al.*, 1996] often require a large number of research platforms and investigators. The ACE are envisioned as a series of international field studies covering a globally representative range of natural and anthropogenically perturbed environments that bring together the platforms and investigators needed to conduct intensive aerosol process and closure studies. To focus the research efforts toward the overall ACE goals, the field experiments are designed with three objectives: (1) to determine the physical, chemical, radiative, and cloud nucleating properties of the major aerosol types and to investigate the relationships between these properties, (2) to quantify the physical and chemical processes controlling the formation, evolution, and fate of the major aerosol types and how these processes affect the number size distribution, the chemical composition, and the

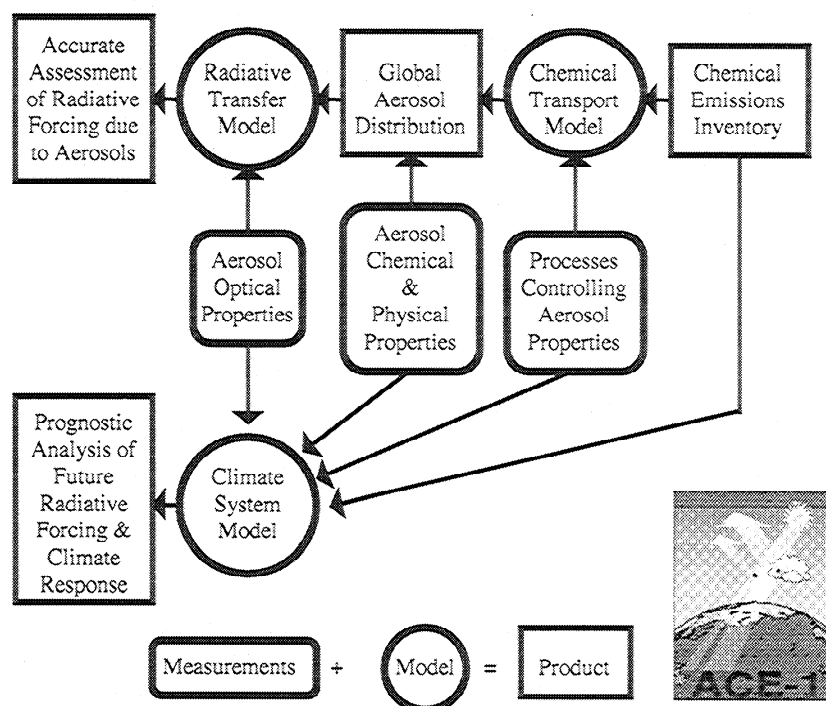


Figure 1. The goals of the ACE are to provide an accurate assessment of radiative forcing due to aerosols and to understand the multiphase atmospheric chemical system sufficiently to be able to provide a prognostic analysis of future radiative forcing and climate response. The measurements of aerosol properties and processes made during ACE 1 are being used to improve chemical transport, radiative transfer, and climate system models to achieve the end product.

radiative and cloud nucleating properties of the particles and, (3) to further develop and test aerosol process, radiative transfer, and regional and global climate models.

3. ACE 1: Study Area

The first ACE took place in the minimally polluted marine atmosphere. The background marine aerosol provided a good starting point for aerosol process studies due to its relative simplicity. The aerosol is largely composed of only two types, a sea-salt component and a non-sea-salt sulfate component. The aerosol distribution in the background marine boundary layer (MBL) is affected by inputs from both the sea surface and the free troposphere. Air-sea exchange of the marine aerosol precursor, dimethylsulfide (DMS), and sea salt affects both the aerosol number and mass size distributions. Convective mixing between the free troposphere and the marine boundary layer, associated with cold frontal passages, cloud pumping, and periods of synoptic-scale subsidence, affects primarily the number size distribution. The background aerosol distributions that result from these processes provide a baseline for comparison with the anthropogenically perturbed aerosol transported from the continent. This baseline is important since radiative forcing, by definition, is an external change in the radiative balance of the Earth. Understanding the background aerosol is necessary to quantify radiative forcing for two reasons. First, radiative forcing is the total radiative effect due to aerosols minus the background radiative effect. Second, since the different aerosol components interact with each other, the total radiative effect is not necessarily the sum of the individual background and anthropogenic components.

The ACE 1 intensive operations took place over the Southern Ocean south of Australia (Figure 2). This site was chosen for the initial experiment for several reasons.

1. This remote southern hemisphere site provided an opportunity to study the natural marine system distant from the northern hemisphere sulfate aerosol.
2. Baseline data collected at Cape Grim and Macquarie Island provided a long-term record of many of the parameters that were measured as part of the intensive experiment. This provided a means by which the intensive experiment could be extrapolated in a broader climatological context.
3. The proximity to Tasmania made the site logistically convenient for ship and aircraft operations.
4. The thin stratocumulus cloud layer, which is present in this area approximately 65% of the time [Jasper and Downey, 1991], was ideal for studying the effects of aerosol particles on cloud properties. Alternately, periods of clear sky were ideal for column closure experiments.
5. The midlatitude oceanic region around Cape Grim provided a rich source of DMS to the atmosphere. The experiment was timed to coincide with the seasonal

maximum DMS concentrations which occur during the November to March time frame [Ayers *et al.*, 1995; Curran *et al.*, this issue].

During the ACE 1 intensive campaign, the synoptic weather pattern was very active with 14 fronts passing through the area and six low-pressure systems distinctly cut off from the prevailing westerlies [Hainsworth *et al.*, this issue]. This intensive cold front activity resulted in below average temperatures, pressures, and rainfall [Hainsworth *et al.*, this issue]. The frequently changing weather patterns provided opportunities to sample background marine, continental, and urban air masses [Whittlestone *et al.*, this issue] and to study the direct and indirect effect of aerosol particles on the Earth's radiative balance. During the month-long campaign, the biological and chemical characteristics of the regional water masses evolved from winter to spring (F. Brian Griffiths *et al.*, manuscript in preparation, 1998) resulting in increasing DMS emissions with time [Bates *et al.*, this issue].

In addition to the month-long intensive campaign, measurements were made on the latitudinal transects down the Pacific in order to make maximum use of the time required to transit the NOAA ship *Discoverer* and NCAR C-130 aircraft to Tasmania (Figure 3). The 31-day transit of *Discoverer* surveyed a variety of oceanic regimes (North Pacific Gyre, equatorial upwelling, South Pacific gyre) and MBL air masses (northern and southern hemisphere westerlies and trade winds) from 48°N to 43°S. The C-130 transit was extended to produce a free tropospheric survey of aerosols and aerosol precursors from 76°N to 59°S.

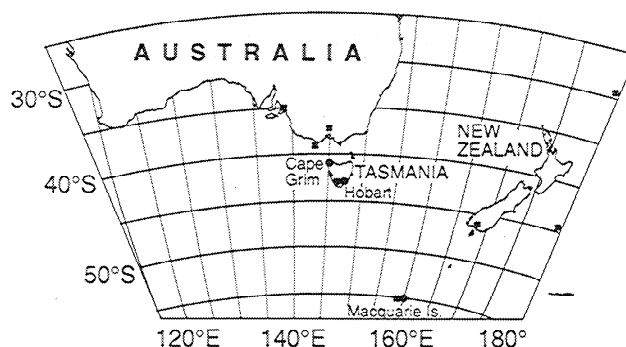


Figure 2. The ACE 1 operations center and aircraft base were located in Hobart, Tasmania. Ground-based measurements were made at Cape Grim and Macquarie Island (solid circles). Aircraft and ship operations took place in the region between 40°–55°S and 135°–160°E. Upper air soundings were conducted from Cape Grim, the R/V *Discoverer*, the R/V *Southern Surveyor*, and the regional meteorological stations (solid squares, Adelaide International Airport, Australia; Chatham Island, New Zealand; Hobart Airport, Australia; Invercargill Airport, New Zealand; Laverton Airport, Australia; Macquarie Island, Australia; Mount Gambier Airport, Australia; Paraparaumu Airport, New Zealand; and Raoul Island, New Zealand).

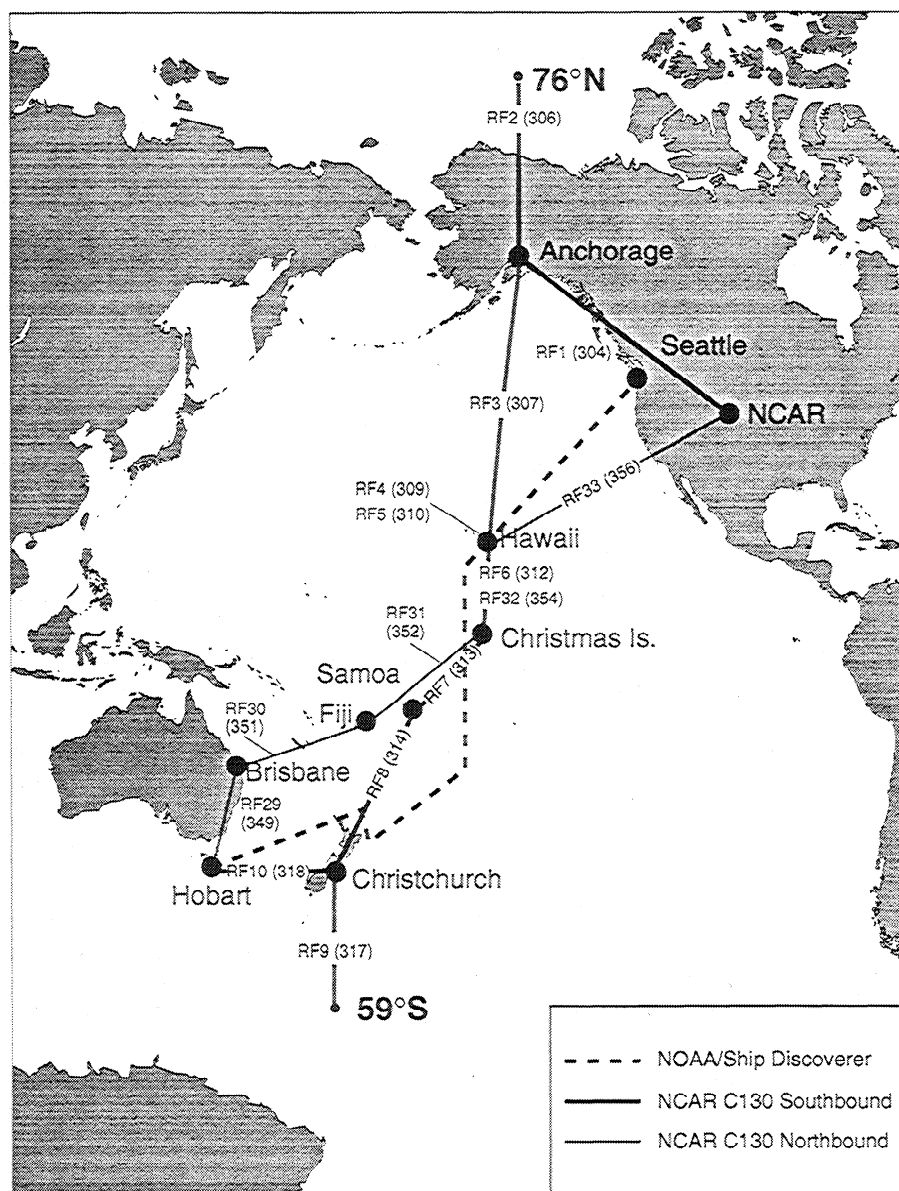


Figure 3. The NCAR C-130 and NOAA R/V *Discoverer* conducted observations between the United States and Australia as part of ACE 1. The aircraft research flights (RF) and day of year (DOY) are shown for each segment of the trip. *Discoverer* departed Seattle on DOY 284, stopped briefly to refuel in Hawaii on DOY 292, and arrived in Hobart on DOY 314.

4. ACE 1 Observations

4.1. Shipboard Measurements

The NOAA R/V *Discoverer* was used to make seawater and atmospheric measurements (Table 1a) during the transit from the United States to Australia (Figure 3) and during the ACE 1 intensive campaign (Figure 4). As a mobile surface station, the shipboard measurements were an essential component of ACE 1. The oceanic DMS measurements provided a means to calculate ocean-atmosphere DMS fluxes which were needed for aerosol process studies [Bates *et al.*, this issue; Mari *et al.*, this issue; Russell *et al.*, this issue; Suhre *et al.*, this issue]. The continuous in situ aerosol measure-

ments that were possible aboard the ship allowed for a more complete characterization of the aerosol chemical, physical, optical, and hygroscopic properties [Bates *et al.*, this issue; Berg *et al.*, this issue; Quinn *et al.*, this issue; Quinn and Coffman, this issue] than was possible from aloft. These data were used for both process and local closure studies. Lower MBL atmospheric gas measurements [De Bruyn *et al.*, this issue; Kok *et al.*, this issue; Moore *et al.*, 1996; T. Carsey *et al.*, manuscript in preparation, 1998], ^{222}Rn [Whit-
tlesstone and Zahorowski, this issue], ozonesondes, and detailed meteorological observations (NCAR Integrated Sounding System (ISS)) provided additional data for aerosol process studies. A satellite receiving station

Table 1a. Discoverer Data Sets

Discoverer Data Sets	Investigator	Institution
<i>Atmospheric Chemical Measurements</i>		
Aerosol mass size distributions, 2 and 7 stage, ions, carbon, gravimetric mass	Patricia Quinn	NOAA/PMEL
Aerosol mass size distributions, high volume, ions	Herman Sievering	University of Colorado
Single particle analysis	Lynn McInnes	NOAA/CMDL
Sulfur dioxide	Eric Saltzman and Warren De Bruyn	University of Miami
Ammonia	Patricia Quinn	NOAA/PMEL
DMS	Timothy Bates	NOAA/PMEL
Ozone (surface and vertical profiles)	James Johnson	NOAA/PMEL
Nonmethane hydrocarbons	Ron Prinn	Massachusetts Institute of Technology
Methyl halides	Robert Moore	Dalhousie University
NO	James Johnson	NOAA/PMEL
NO, NO ₂ , NO _y	Tom Carsey	NOAA/AOML
Radon	James Johnson and Stewart Whittlestone	NOAA/PMEL ANSTO
CO	James Johnson	NOAA/PMEL
pCO ₂	Richard Feely	NOAA/PMEL
<i>Aerosol Physical and Optical Measurements</i>		
Particle number size distributions (5–5000 nm diameter)	Timothy Bates	NOAA/PMEL
Aerosol hygroscopic growth (H-TDMA)	Erik Swietlicki	Lund University
Aerosol volatility (T-TDMA)	Alfred Wiedensohler	Institute for Tropospheric Research
Aerosol light scattering and absorption	Patricia Quinn	NOAA/PMEL
Aerosol optical depth	Patricia Quinn	NOAA/PMEL
Satellite observations	Phillip Durkee	Naval Postgraduate School
<i>Seawater Measurements</i>		
DMS	Timothy Bates	NOAA/PMEL
DMSP/DMSO	Ron Kiene	University of South Alabama
Ammonia and pH	Patricia Quinn	NOAA/PMEL
Nonmethane hydrocarbons	Ron Prinn	Massachusetts Institute of Technology
Methyl halides	Robert Moore	Dalhousie University
POC, PON, POS, DOC, DON	Yoshimi Suzuki	Shizuoka University
SST, salinity, nitrate	Timothy Bates	NOAA/PMEL
CTD profiles	James Johnson	NOAA/PMEL
<i>Meteorological Measurements</i>		
Surface meteorological data	James Johnson	NOAA/PMEL
Integrated sounding system (ISS) balloon, profiler, RASS data		NCAR
Spectroradiometer (300–850 nm)	James Johnson	NOAA/PMEL
Cloud (sky camera) images	James Johnson	NOAA/PMEL

aboard the ship during the transit and at the Hobart operations center during the intensive operations provided a means to relate the shipboard in situ measurements with column-integrated aerosol optical depth estimates from shipboard sunphotometer and NOAA advanced very high resolution radiometer (AVHRR) measurements (P. Durkee et al., manuscript in preparation, 1998). Finally, the ship provided surface support for the Lagrangian (S. Businger et al., manuscript in preparation, 1998) and column closure experiments.

Air was sampled from a number of locations on the ship (Figure 5). Most inlets were located near the top of the aerosol mast or directly behind this location on the ship's flying bridge. Aerosol particles were sampled at 18 m above sea level (asl) through the heated mast. The mast extended 6 m above the aerosol measurement container and was capped with a rotating inlet nozzle that was positioned into the relative wind. Air was pulled through this 5 cm diameter inlet nozzle at $1 \text{ m}^3 \text{ min}^{-1}$ and down the 20 cm diameter mast. The lower 1.5 m

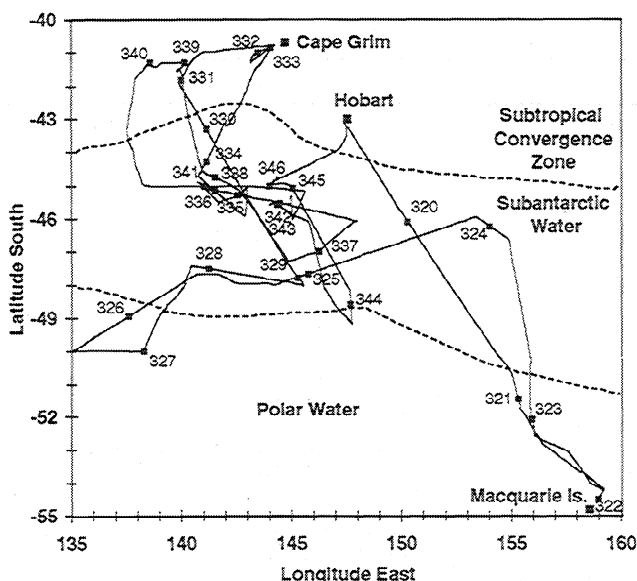


Figure 4. *Discoverer* cruise track during the intensive operations of ACE 1. The numbers (day of year in 1995) represent the ship's position at the beginning of each day (UTC). The three surface water masses are defined by salinity with subtropical convergence zone salinities >34.8 practical salinity unit (psu), Subantarctic water salinities >34.2 psu but <34.8 psu, and polar water salinities <34.2 psu.

of the mast were heated to dry the aerosol to an RH of $<50\%$. Fifteen 1.9 cm diameter tubes extending into this heated zone were used to isokinetically subsample the air stream for the various aerosol sizing instruments and impactors at flows of 30 l min^{-1} .

Seawater was sampled from the ship's clean seawater inlet at the bow of the ship, nominally 5 m below the sea surface (Figure 5). Water samples were also collected for a limited number of species using the ship's Seabird conductivity/temperature/depth (CTD)

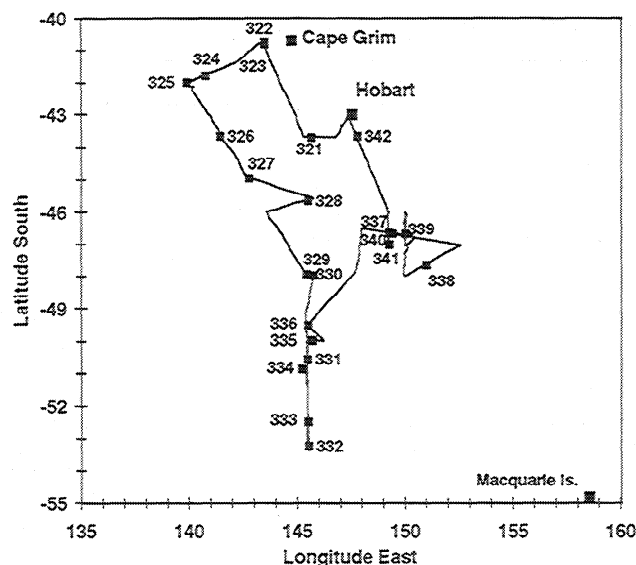


Figure 6. *Southern Surveyor* cruise track during ACE 1. The numbers (day of year in 1995) represent the ship's position at the beginning of each day (UTC).

rosette sampler. Fifty-four CTD deployments were conducted at 28 sampling stations roughly evenly spaced along the transit from Seattle to Hobart. Eight CTD deployments were conducted from *Discoverer* during the ACE 1 intensive covering the three main water masses in the region (Figure 4).

Shipboard measurements were also conducted during ACE 1 aboard the Australian R/V *Southern Surveyor* (Figure 6 and Table 1b). Although the primary objective of the R/V *Southern Surveyor* was to study ocean carbon cycling as part of the Joint Global Ocean Flux Study (JGOFS), the chemical, biological, and ocean optical measurements made a valuable addition to the ACE 1 data set and were particularly im-

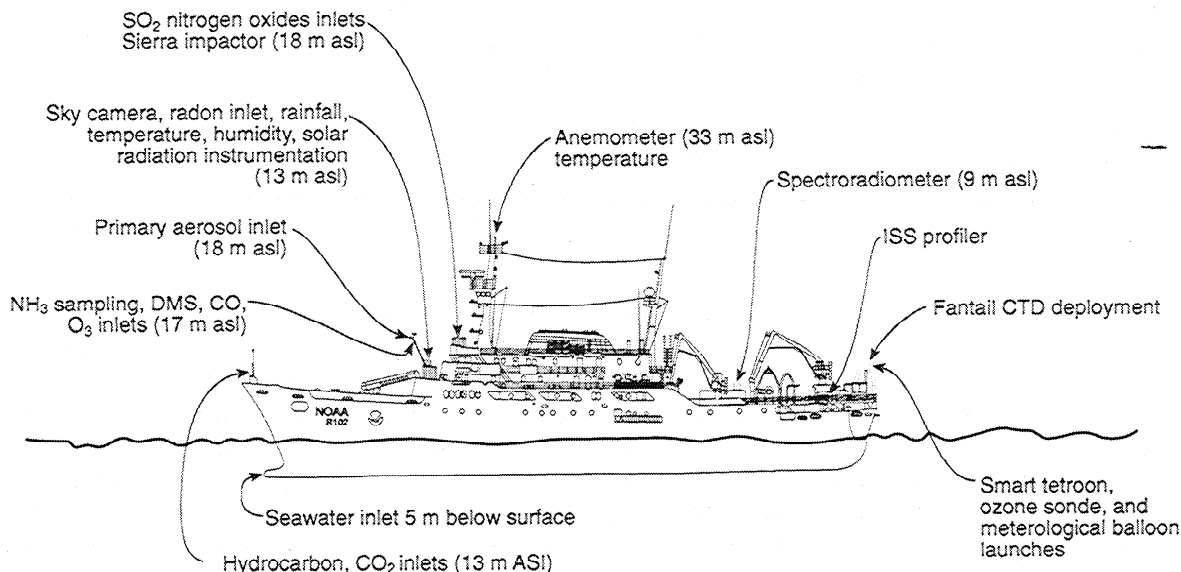


Figure 5. Sampling and instrumentation locations (meters above sea level) aboard *Discoverer*.

Table 1b. Southern Surveyor Data Sets

Southern Surveyor Data Sets	Investigator	Institution
<i>Underway Data</i>		
Nutrients, fluorescence	Brian Griffiths	CSIRO Division of Marine Research
fCO ₂	Bronte Tilbrook	CSIRO Division of Marine Research
POC	Tom Trull	Antarctic CRC, University of Tasmania
Ammonia	Lesley Clementson	CSIRO Division of Marine Research
Acoustic sampling	Rudy Kloser	CSIRO Division of Marine Research
Fast-repetition rate fluorometry	Rick Greene	Texas A&M University
DMS/DMSP	Graham Jones and Mark Curran	James Cook University and Antarctic CRC, University of Tasmania
Radon	Stewart Whittlestone	ANSTO
Aquashuttle	Don McKenzie	CSIRO Division of Marine Research
<i>Vertical/CTD</i>		
Particle size analysis	Pru Bonham	CSIRO Division of Marine Research
HPLC chlorophyll and spectral absorbance	Lesley Clementson	CSIRO Division of Marine Research
Sediment trap analyses	Lesley Clementson	CSIRO Division of Marine Research
Filtered nutrients	Lesley Clementson	CSIRO Division of Marine Research
DMS/DMPS	Graham Jones and Mark Curran	James Cook University and Antarctic CRC, University of Tasmania
Cell cycling/phytoplankton growth rates	Rick Greene	Texas A&M University
Vertical fast-repetition rate fluorometry	Rick Greene	Texas A&M University
Phytoplankton species	Rick Greene	Texas A&M University
P-I studies/production modeling	Brian Griffiths and Pru Bonham	CSIRO Division of Marine Research
¹⁵ N uptake	Brian Griffiths	CSIRO Division of Marine Research
Grazing dilution	Brian Griffiths	CSIRO Division of Marine Research
CTD, nutrients	Brian Griffiths	CSIRO Division of Marine Research
Fluorescence and PAR profiles	Brian Griffiths	CSIRO Division of Marine Research
Ammonia	Lesley Clementson	CSIRO Division of Marine Research
Optics	John Parslow and Don McKenzie	CSIRO Division of Marine Research
Alkalinity/pCO ₂ / ¹³ C/DIC	Bronte Tilbrook	CSIRO Division of Marine Research
DOC/POC/ ¹³ C	Tom Trull	Antarctic CRC, University of Tasmania

portant in assessing the processes controlling seawater DMS concentrations [Jones *et al.*, this issue]. Atmospheric ²²²Rn [Whittlestone and Zahorowski, this issue] and radiosonde measurements (Figure 7) aboard *Southern Surveyor* also helped to characterize the air masses in the ACE 1 study area.

4.2. Aircraft Measurements

The NCAR C-130 was used to make a comprehensive suite of chemical and physical measurements at a variety of altitudes. It was configured to make eddy correlation measurements of ozone, sensible heat, water, and momentum, in addition to aerosol microphysics and chemistry, cloud properties, radiation, and the concentrations of oxidants, sulfur gases, hydrocarbons, and halocarbons. It also carried NCAR's Scanning Aerosol Backscatter Lidar (SABL) for observing aerosols above and below the aircraft and selecting altitudes for in situ sampling legs. Sampling profiles were designed to observe boundary layer processes, the potential for upper tropospheric aerosol nucleation, and vertical distributions of aerosols and radiation.

The location of the major instrumentation (Table 1c) aboard the C-130 is shown in Figure 8. Most of the aerosol instruments received air from the community aerosol inlet (CAI). This shrouded inlet sampled ambient air isokinetically, then decelerated it to about 10 m/s in three linear diffuser sections before remov-

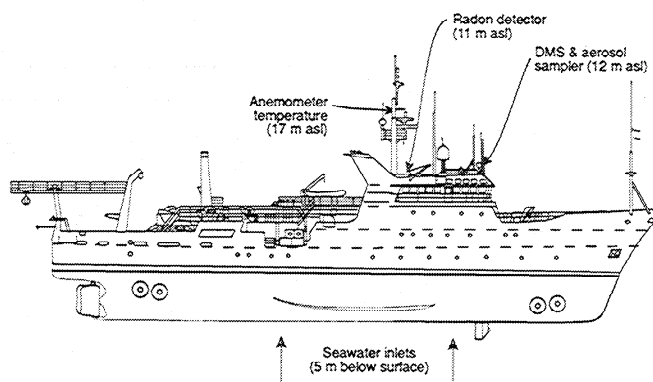


Figure 7. Sampling and instrumentation locations (meters above sea level) aboard *Southern Surveyor*.

Table 1c. C-130 Data Sets

C-130 Data Sets	Investigator	Institution
<i>Atmospheric Chemical Measurements</i>		
Aerosol ionic mass size distributions, 2 stage	Barry Huebert	University of Hawaii
Aerosol organic speciation	John Seinfeld	California Institute of Technology
Ammonia and NO	John Bradshaw	Georgia Institute of Technology
Single-particle analysis	Jim Anderson	Arizona State University
Gas phase OH, H ₂ SO ₄ , and MSA	Fred Eisele	NCAR
DMS and SO ₂	Alan Bandy and Donald Thornton	Drexel University
Ozone (UV and NO chemiluminescence)	Greg Kok	NCAR
Nonmethane hydrocarbons, methyl halides	Don Blake and Sherry Rowland	University of California, Irvine
H ₂ O ₂ , CO, CO ₂	Greg Kok	NCAR
<i>Aerosol Physical and Optical Measurements</i>		
Particle number size distributions (3–500 nm diameter)	Lynn Russell and John Seinfeld	Princeton University and California Institute of Technology
Nanoparticle (3–10 nm diameter) spectra by UCNC pulse height analysis	Peter McMurry and Rodney Weber	University of Minnesota and Brookhaven National Laboratory
CN, UCN, RCN (heated/ambient CNCs)	Tony Clarke	University of Hawaii
Inferred aerosol composition versus size (heated/ambient OPC)	Tony Clarke	University of Hawaii
Aerosol light scattering and absorption	Tony Clarke	University of Hawaii
Cloud condensation nuclei (CCN)	Jim Hudson	Desert Research Institute
Upwelling and downwelling radiation	Francisco Valero	University of California, San Diego
Upwelling radiance (ocean color)	John Porter	University of Hawaii
Aerosol vertical profiles (LIDAR)	Larry Radke and Bruce Morley	NCAR
Particles >0.1 μ m, ASASP, MASP, OPCs	Darrel Baumgardner	NCAR
<i>Ancillary Measurements</i>		
Balloon location	Steve Businger	University of Hawaii
<i>Meteorological Measurements</i>		
RH, temperature, pressure, altitude, location	RAF	NCAR

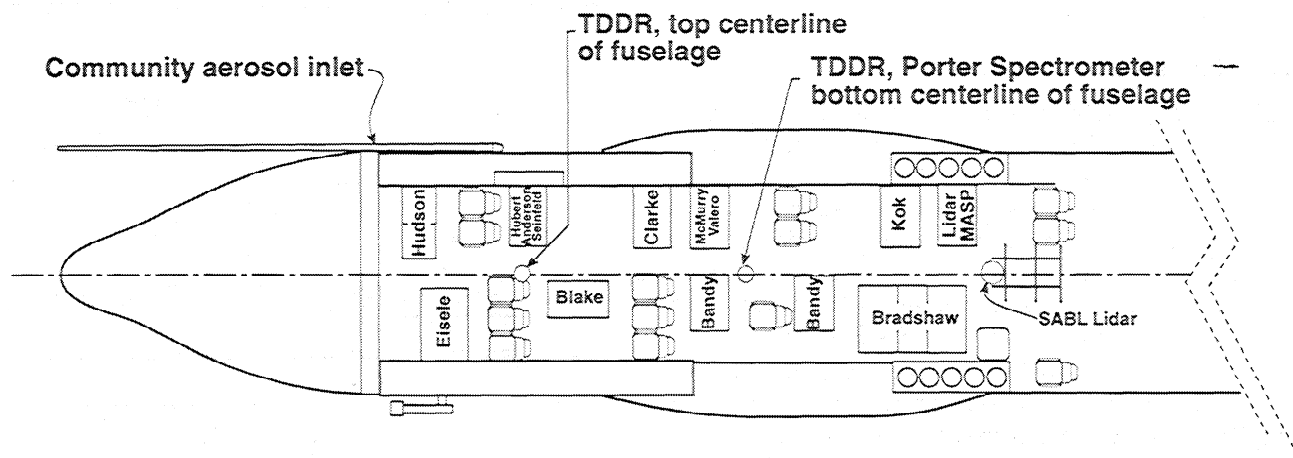


Figure 8. Sampling and instrumentation locations aboard the NCAR C-130. The community aerosol inlet (CAI) is shown on the starboard side of the aircraft.

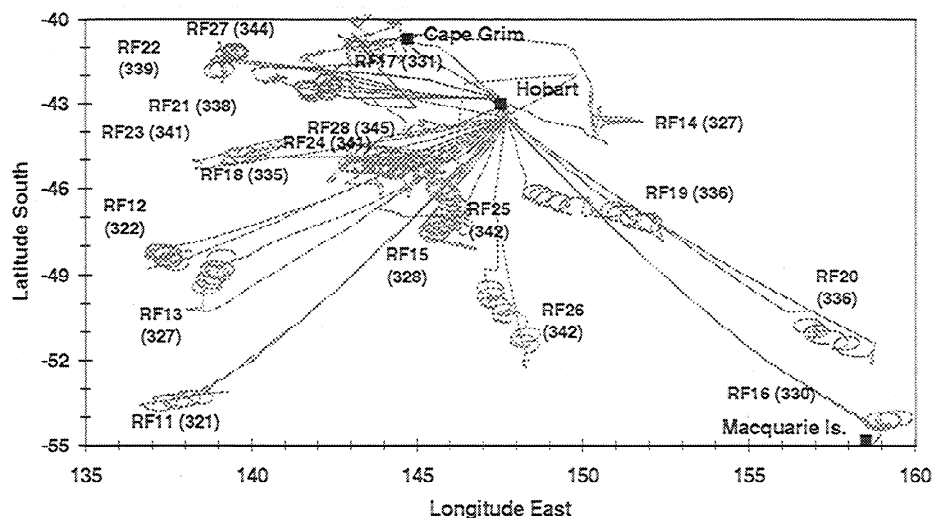


Figure 9. C-130 flight tracks during ACE 1. The research flight (RF) number and day of year at takeoff (UTC) are shown for each flight.

ing part of the flow at a sampling plane and bringing that air into the fuselage. The CAI was intended to minimize the loss of aerosols on inlet systems and to ensure that any losses would at least be similar for all the instruments being used for local closure experiments. The performance of the CAI has been further tested in a post-ACE 1 experiment, CAINE-2 (Community Aerosol Inlet Experiment), and will be reported at a later date.

The C-130 flew a total of 288 flight hours during ACE 1 in 33 research flights. Of this total, 162 hours were flown out of Hobart, between November 15 and December 14, 1995 (Figure 9). Seventy-one hours were devoted to Lagrangian experiments, 19 hours to studying nucleation in the outflow of convective clouds, 18 to looking at the dynamics of stratocumulus cloud systems, 16 to column closure, 9.2 to a Macquarie Island survey, and 29 to a variety of other objectives.

There was no "ferry" time for the C-130 in the usual sense, since the aircraft conducted a full suite of observations on the 126 hours of transects to and from Hobart. The southbound transect (RF1–RF10) spanned the latitude range of 76°N to 59°S (Figure 3) and included two flights (RF4, sulfur evolution and RF5, column closure) in the Kilauea volcano plume. The plane flew first to Anchorage, Alaska (RF1), from which it flew a loop as far north as possible (RF2), including two legs low over the ice of the Beaufort Sea. It then stepped down the central Pacific, stopping in Hawaii (RF3), Kiribati (RF6), Western Samoa (RF7), Christchurch, New Zealand (RF8), and Hobart (RF10). From Christchurch it made another loop, this time as far south as possible (RF9). The northbound transect took a more direct route, with stops in Brisbane (RF29), Fiji (RF30), Kiribati (RF31), Hawaii (RF32), and Colorado (RF33).

One of the transect objectives was to see whether or not ultrafine nuclei (and thus nucleation) are largely

confined to the regions where recently scavenged air is being expelled from upwelling air. The flights included cruise legs at about 6 km altitude, with a few boundary layer legs on most flights and vertical profiles near Kiribati, Brisbane, and Fiji. Another major scientific objective of the transects was deriving latitudinal profiles of all the gases, aerosols, and optical properties that were measured on the C-130 [Baumgardner and Clarke, this issue; Hudson *et al.*, this issue; Kok *et al.*, this issue].

Seventy-one flight hours were used for two Lagrangian experiments. In each Lagrangian, the C-130 made multiple flights to sample an air mass which had been marked by balloons launched from the R/V *Discoverer* (S. Businger *et al.*, manuscript in preparation, 1998). Lagrangian A began prior to sunrise and consisted of three flights, RF18, RF19, and RF20. The first two flights were oriented a few kilometers north of a single balloon, but its radio signal could not be located during RF20. Stacks were flown in circular patterns at a variety of altitudes within and just above the boundary layer. Lagrangian B consisted of a pre-Lagrangian survey flight (RF23) to assess the source of air behind a weak frontal system, followed by three flights (RF24–26) in that air. In this case all three flights were made in the vicinity of three balloons, which were still operational when their distance from Hobart finally required the airborne observations of that air mass be terminated. The evolution of species in a Lagrangian reference frame is the subject of numerous papers in this issue [Huebert *et al.*, this issue; Mari *et al.*, this issue; Russell *et al.*, this issue; Suhre *et al.*, this issue; Q. Wang *et al.*, unpublished manuscript, 1998].

C-130 flights addressed a wide variety of related objectives in the remaining flights from Hobart. Studies of sulfur chemistry at low temperatures and around sunrise motivated RF11 and RF21, respectively. Stratocumulus dynamics was the focus of RF12 and RF28 [Boers

et al., this issue; *Boers and Krummel*, this issue]. Measurements in and around clouds also provided data to quantify photodissociation and UV-radiative transfer in a cloudy atmosphere [*Matthijsen et al.*, this issue]. Profiles in the vicinity of R/V *Discoverer* and intercomparisons between ship and airborne concentration measurements and flux methods were the focus of RF15, RF22, and RF28. Profiles above and upwind of Cape Grim were made on RF14 and RF27. Flights RF14, RF17, and RF27 included studies of new particle production in the outflow regions of cumulus clouds [*Clarke et al.*, this issue]. The last of these clearly showed that photochemical processes in cloud outflow can create new particles. A long transit to Macquarie Island (RF16) provided an opportunity to observe the influence of ammonia vapor on homogeneous nucleation [*Weber et al.*, this issue]. Aerosol column closure experiments were conducted on RF5, RF14, and RF27. Details of many of these flights can be found in the papers that follow.

4.3. Ground-Based Measurements

Measurements at ground stations on Macquarie Island and Cape Grim comprised an integral part of the ACE 1 strategy. These stations provided the opportunity for continuous records of aerosol and related parameters uncomplicated by varying geographic location.

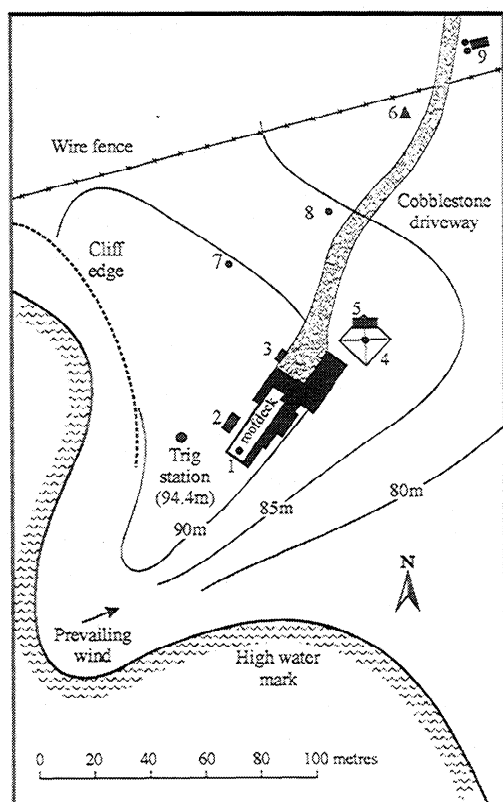


Figure 10. Plan view of the Cape Grim Baseline Station showing (1) 10 m inlet, (2) Van laboratory, (3) liquid water radiometer, (4) 74 m mast, (5) PALMS laboratory, (6) ISS surface mast, (7) Stevenson screen, (8) station exhaust, and (9) ISS laboratory.

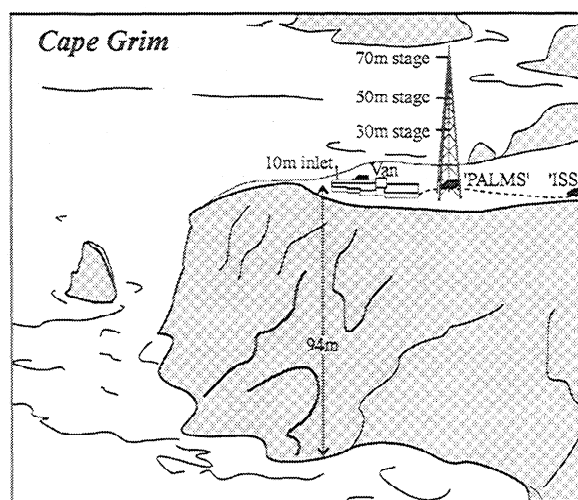


Figure 11. Sampling locations at Cape Grim depicted schematically in Figure 10, from a perspective approximately SE of the station.

The data obtained are well suited as input to regional models; they provide an overall context for the experiment in relation to established climatologies; and they provide an opportunity to carry out local closure studies over extended periods in conditions varying from clean maritime to urban-influenced-continental and biomass burning.

Cape Grim (40.7°S, 144.7°E), the site of Australia's Baseline Atmospheric Pollution Program has a long record of several aerosol properties. This includes, for example, concentrations of CN (with $D_p > 3$ nm) since 1976, CCN (0.25–1.2% supersaturation) since 1981, and soluble chemical species since 1976 [*Ayers et al.*, 1991; *Gras*, 1995]. The station is located 94 m above sea level, on a steep bluff overlooking the Southern Ocean (Figures 10 and 11). The majority of the instrumentation used during ACE 1 was located in the main observatory laboratory and utilized the community 10-m inlet. This inlet consisted of a straight stainless-steel stack 150 mm in diameter with a flow in the stack maintained at a Reynolds number around 2300. The main flow was subsampled under close to isokinetic conditions using a series of tapered inlets arranged concentrically at approximately 1 m above the base of the stack. Humidity controls were installed to provide operation at a maximum of 50% RH for size-selective inlets.

Sampling was also conducted on the main laboratory roof deck, from the tower at both the 30 m and 50 m levels (Figure 11), and in a temporary laboratory (PALMS) located directly underneath the tower (Figure 10). A 100-mm diameter vertical stainless-steel sample stack running up the center of the tower was used to supply sample air from the 50-m level to the temporary laboratory at the base of the tower. Sampling from this stack at the base of the tower was approximately isokinetic. Temporary laboratories were also located on the western side of the main station

(VAN) and about 100 m north of the station (ISS). Instrumentation used during ACE 1 and the various inlet locations are listed in Table 1d.

As in the shipboard measurements, the continuous in situ aerosol measurements at Cape Grim allowed for a more complete characterization of the aerosol prop-

erties than was possible from aloft [Middlebrook *et al.*, this issue; Murphy *et al.*, this issue, 1997, 1998; J. Anderson *et al.*, manuscript in preparation, 1998; Carrico *et al.*, this issue; Covert *et al.*, this issue; J. Gras *et al.*, manuscript in preparation, 1998]. The data were used for a number of local closure studies, for example, mass,

Table 1d. Cape Grim and Hobart Data Sets

Cape Grim and Hobart Data Sets	Investigator	Institution
<i>Atmospheric Chemical Measurements</i>		
Aerosol mass size distributions, 11 stages, weekly, ions (RF)	Greg Ayers and Jill Cainey	CSIRO
Aerosol mass size distributions, 2 stages, ions (PL)	Greg Ayers and Jill Cainey	CSIRO
Aerosol mass size distributions, 9 stages, ions, organic, gravimetric (ML)	Barry Huebert	University of Hawaii
Aerosol mass size distributions, high volume, 6 stages, ions (T50)	Herman Sievering	University of Colorado
Aerosol total mass, high volume, ions (T30)	Herman Sievering	University of Colorado
Aerosol total mass, 3 hourly, ions (T30)	Barry Huebert	University of Hawaii
Aerosol gravimetric mass, submicron (ML)	John Gras	CSIRO
Single-particle SEM/EDXA analysis (T50)	Jim Anderson	Arizona State University
Single-particle mass spectrometry (PL)	Dan Murphy and Ann Middlebrook	NOAA/AL
Ammonia (Van)	Pai-Yei Whung	NOAA/AOML
DMS (ML)	John Ivey	AGAL
Ozone (ML)	Ian Galbally	CSIRO
Nonmethane hydrocarbons and halocarbons (ML)	Ian Galbally	CSIRO
Peroxides (Van)	Greg Ayers	CSIRO
NO _x	Ian Galbally	CSIRO
Radon (ML)	Stewart Whittlestone	ANSTO
CO and pCO ₂	Paul Steele	CSIRO
<i>Aerosol Physical and Optical Measurements</i>		
Particle number size distributions (5–5000 nm diameter) (ML)	Alfred Wiedensohler	Institute for Tropospheric Research
Particle number size distributions (3–3000 nm diameter) (ML)	John Gras	CSIRO
Particle number size distributions (300–10000 nm diameter) (PL)	Dan Murphy and Ann Middlebrook	NOAA/AL
Aerosol hygroscopic growth (H-TDMA) (ML)	Dave Covert	University of Washington
Aerosol volatility (T-TDMA)	Alfred Wiedensohler	Institute for Tropospheric Research
Aerosol light scattering (40 and 85% RH) (ML)	Mark Rood	University of Illinois
Aerosol light absorption (ML)	John Gras	CSIRO
CCN at 0.3, 0.5, and 1.2% supersaturation (ML)	John Gras	CSIRO
CCN (PL*)	Jim Hudson	Desert Research Institute
CN ($D_p > 5$ nm) (PL*)	Jim Hudson	Desert Research Institute
CN ($D_p > 3, 5, \text{ and } 10$ nm) 5 counters (ML)	John Gras and Dave Covert	CSIRO and University of Washington
Aerosol optical depth (RF)		Bureau of Meteorology
Aerosol optical depth (Hobart)		Bureau of Meteorology
Satellite observations (Hobart)	Philip Durkee	Naval Postgraduate School
<i>Meteorological Measurements</i>		
Surface meteorological data		Bureau of Meteorology
Integrated sounding system (ISS) balloon, profiler, RASS data		NCAR

ML, Main laboratory, 10-m inlet; PL, PALMS laboratory, 50-m inlet; T30, tower 30-m level; T50, tower 50-m level; Van, Van Laboratory; ISS, ISS Laboratory.

*Limited period of operation.

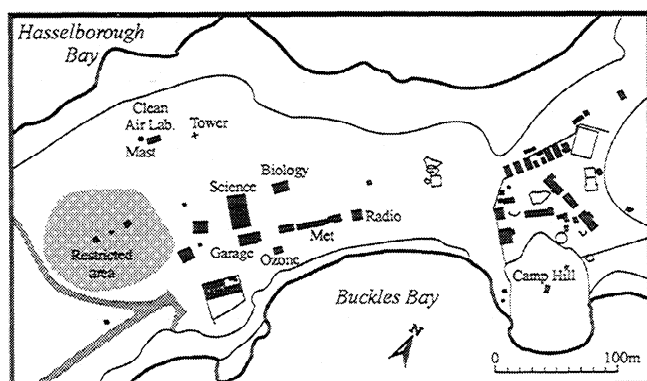


Figure 12. Plan view of the Macquarie Island Research Station.

scattering coefficient, hygroscopic growth, CCN activity, and in process studies. The supporting gas measurements (DMS, ^{222}Rn [Whittlestone and Zahorowski, this issue], NO_x , NH_3 , peroxides, O_3), and detailed meteorology observations (continuous surface observations, Doppler wind profiler, radio acoustic sounding system, and balloon soundings), all contributed to a comprehensive understanding of the processes controlling aerosol properties.

Macquarie Island (54.5°S , 159.0°E) is a Subantarctic island approximately midway between Tasmania and the Antarctic continent (Figure 2). The clean-air laboratory on Macquarie Island is located on a sandy isthmus about 100 m upwind of the station complex (Fig-

ure 12). Limited observations of aerosol properties have been obtained at Macquarie Island since 1986, including total CN concentration and some soluble species [Gras, 1991]. During ACE 1, aerosol studies focused on microphysical characterization [Brechtel *et al.*, this issue; Kreidenweis *et al.*, this issue] and CCN local closure. Most equipment was located in the clean-air laboratory (CAL). Sampling was conducted from 5 m (12 m asl) and 10 m (17 m asl) inlets located near the SW corner of the laboratory, from instruments attached to a 10-m tower adjacent to the laboratory or from a separate free-standing walk-up 10-m tower that was installed north of the laboratory for ACE 1 (Figure 12). Instrumentation, locations, and investigators are listed in Table 1e.

4.4. Satellite Observations

Observations from satellite platforms provided valuable input to mission planning as well as post-experiment analysis. Real-time receipt of satellite imagery provided the regional characteristics of aerosol and cloud properties as well as sea surface temperature and basic meteorological information. The satellite analysis was used to identify regions and features that met the ACE 1 objectives. The ship and aircraft operations were designed to be flexible to changes observed in the satellite data. NOAA AVHRR data were collected in real time aboard the *Discoverer* on the transit from Seattle to Hobart and at the operations center during the aircraft/ship studies around Tasmania. Geostationary imagery from the Japanese GMS satellite provided im-

Table 1e. Macquarie Island Data Sets

Macquarie Island Data Sets	Investigator	Institution
<i>Atmospheric Chemical Measurements</i>		
Aerosol mass size distributions, 2 stage, ions, (CAL)	Chad Dick	Bureau of Meteorology
Aerosol mass size distributions, high volume, ions, (TWR)	Neil Tindale	Texas A&M University
DMS (CAL)	Hilton Swan	AGAL
Radon (CAL)	Stewart Whittlestone	ANSTO
<i>Aerosol Physical and Optical Measurements</i>		
Particle number size distributions (120–3000 nm diameter) (CAL)	Mike Harvey and Georgina Sturrock	NIWA
Particle number size distributions (10–200 nm diameter) (CAL)	Sonia Kreidenweis and Fred Brechtel	Colorado State University
Total particle number concentration ($D_p > 3$ and $D_p > 10$ nm) (CAL)	John Gras, Sonia Kreidenweis and Fred Brechtel	CSIRO and Colorado State University
Cloud condensation nuclei (CCN) (CAL)	Mike Harvey and Georgina Sturrock	NIWA
<i>Meteorological Measurements</i>		
Surface meteorological data and balloon soundings		Bureau of Meteorology

CAL, clean air lab; TWR, TAMU tower; AGAL, Australian Government Analytical Laboratories; ANSTO, Australian Nuclear Science and Technology Organisation; NIWA, National Institute of Water and Atmospheric Research Ltd.

portant temporal information about cloud motion and synoptic systems for forecasting and mission planning. Real-time analysis of these observations and updates to mission plans were routinely radioed to the ship and aircraft, often within minutes of receipt of satellite data. This capability maximized mission execution by assuring the best possible orientation of the ship and aircraft operations. In post-experiment studies, analysis of satellite observations helped to place in situ measurements in the context of the regional environment. The AVHRR instrument measured radiance at 1-km resolution over a 2500-km-wide swath on each pass of the satellite. Analysis of NOAA AVHRR measurements provided estimates of aerosol optical depth (P. A. Durkee et al., manuscript in preparation, 1998), aerosol size characteristics, cloud reflectance at visible ($0.63\ \mu\text{m}$) and near-infrared ($3.7\ \mu\text{m}$) wavelengths, cloud amount, cloud top temperature [Bates et al., this issue], and sea surface temperature. These observations provided a means to compare the characteristics observed in ship, aircraft, and surface data to properties of the regional-scale environment.

ACE 1 also provided several opportunities to validate satellite retrieval techniques with in situ observations. Three clear-column closure experiments were conducted to characterize the properties of the atmospheric column that affect retrieval of aerosol properties. Sunphotometer measurements on board *Discoverer* also provided validation of satellite optical depth retrievals during the entire ACE 1 time period.

4.5. Intercomparisons of Measurements on Various ACE 1 Platforms

The value of a multiplatform experiment depends critically on being able to compare data from many different investigators and their many instruments. ACE 1 included both laboratory and field intercomparisons of all the major sampling systems. Laboratory workshops, which were designed to resolve questions about calibrations and techniques without uncertainties due to platforms and slightly differing field locations, were run before the field campaign. Pre-ACE 1 workshops intercompared the size dependent counting efficiencies of the 26 condensation particle counters used in ACE 1 [Wiedensohler et al., 1997], tested particle charging and transmission efficiencies of aerosol charge neutralizers [Covert et al., 1997], examined differential mobility analyzer transfer functions [Birmili et al., 1997], intercompared the CCN counters used in ACE 1, and intercompared ion chromatography standards used in analyzing ACE 1 samples. Field comparisons were conducted by sampling or sensing from colocated platforms throughout the field intensive portion of ACE 1.

5. ACE 1 Research Strategy

The ACE scientific objectives were addressed in several activities during the intensive measurement campaign and transit flights and cruise between the United

States and Australia. The activities focused on six research questions. A critical part of aerosol radiative forcing calculations involves relating the size-dependent chemical composition of aerosols to their radiative impact. In models, this module makes the critical link between anthropogenic changes in the chemical composition of the atmosphere and the associated impact on the Earth's radiation budget. The first two questions addressed objective 1 and were meant to provide data to test and validate aerosol/climate models on local and column-integrated scales.

Process studies are needed to evaluate the factors controlling the sources, formation, and fate of aerosols and the way these processes affect the number size distribution, chemical composition, and radiative and cloud nucleating properties of the particles. The next three questions addressed objective 2 and related to models of the natural biogeochemical processes that create atmospheric aerosols and the processes that control the aerosol population. Finally, the sixth question, which addressed objective 3, integrated the closure experiments and process studies into models on a variety of scales.

5.1. In-Situ Characterization: Closure Experiments

Objective 1 is to determine the physical, chemical, radiative, and cloud nucleating properties of the major aerosol types and to investigate the relationships between these properties. Simultaneous measurements of aerosol chemical, physical, radiative, and cloud nucleating properties are needed to develop and test model calculations of aerosol radiative effects and to refine and validate the algorithms used to interpret satellite observations of the aerosol [Penner et al., 1994]. A key concept behind integrating models and measurements is the closure experiment [Quinn et al., 1996]. In such an experiment an overdetermined set of observations is obtained, and the measured value of a dependent variable, for example, measured light scattering by aerosols, is compared with the value that is calculated from the measured values of the independent variables, for example, measured aerosol chemical and physical properties, using an appropriate model, for example, Mie scattering model. The outcome of a closure experiment provides a direct evaluation of the combined uncertainty of the model and measurements. If there is agreement between the measured and calculated values within the accepted level of uncertainty, the model may be a suitable representation of the observed system and appropriate for use as a component in other higher-order models. Poor agreement indicates that there are problems in the model or measurements that must be corrected before proceeding further. The closure experiments conducted during ACE 1 fall into two broad classes, local closure and column closure.

A local closure experiment is one in which all measurements are made at a single location and time. Several local closure experiments are required to address

uncertainties in the estimated climate forcing by aerosols. The unknowns include the aerosol mass scattering efficiency, aerosol hemispheric backscatter fraction, fractional increase in aerosol scattering efficiency due to hygroscopic growth, and parameters affecting aerosol activation [Penner *et al.*, 1994]. All of these experiments relate to one specific question: question 1, Can the measured physical and chemical properties of the aerosol be used to predict the radiative and cloud nucleating properties of that same aerosol?

The data collected during ACE 1 are being used in six local closure experiments. The first local closure experiment compared aerosol mass as a function of particle size derived from the number size distribution, chemical analysis of aerosol species, and gravimetric analysis. Key measurements for each local closure experiment include the aerosol number concentration and chemical composition as a function of particle size. It is important, therefore, to validate these measurements and to estimate their uncertainty. The mass closure analysis addressed the internal consistency of these measurements; that is, did the chemically analyzed mass account for the total aerosol mass? Was the mass derived from the aerosol chemical size distribution consistent with that from the aerosol number size distribution? Results from this study conducted aboard *Discoverer* showed closure between the gravimetric mass, the chemically analyzed ionic mass, and the calculated number-size distribution mass [Quinn and Coffman, this issue]. In contrast, the aerosol measured at Cape Grim appeared to have a significant organic component [Huebert *et al.*, this issue; Middlebrook *et al.*, this issue].

The second local closure experiment compared the measured hygroscopic response of the aerosol size to changes in relative humidity (RH) and the hygroscopic response calculated from the measured aerosol number and chemical mass size distributions, RH, and published functional relationships between chemical composition and water uptake. This intercomparison added an additional step of complexity to intercomparison 1 by varying the total water content of the aerosol and addressed the ability to parameterize the response of ambient aerosol to changes in RH. Results from this study will be reported at a later date.

The third local closure experiment compared measured aerosol scattering and calculated aerosol scattering derived from Mie theory applied to measured number and chemical mass size distributions. This intercomparison directly addressed the uncertainty in determining the aerosol/climate parameters of aerosol mass scattering efficiency and aerosol hemispheric backscatter fraction. Scattering and backscattering of visible light by particles was measured directly with an integrating nephelometer. These direct measurements of light scattering properties were compared with independent predictions from the aerosol number and chemical mass size distributions, some simplifying assumptions (e.g., spherical particle shape, an externally mixed

aerosol, particle refractive index, and density), and Mie theory. Results from this study conducted aboard *Discoverer* showed closure for the submicron size range but a lack of closure for the super-micron particles [Quinn and Coffman, this issue]. The results also show that sea salt was the dominant chemical component controlling aerosol optical properties [Murphy *et al.*, 1998; Quinn *et al.*, this issue].

The fourth local closure experiment compared the measured increase in aerosol scattering due to hygroscopic growth and the calculated increase based on measured number and chemical mass size distributions, RH, and published functional relationships between particle chemical composition and water uptake. This intercomparison added an additional step of complexity to intercomparison 3 by varying the total water content of the aerosol. It addressed the aerosol/climate parameter of fractional increase in aerosol scattering efficiency due to hygroscopic growth. Two nephelometers were operated in parallel, to measure directly the scattering and backscattering of visible light by particles at a low reference RH (10%) and at two higher RH levels. The increase in aerosol scattering due to the aerosol hygroscopic growth was independently calculated using the data from intercomparisons 2 and 3. Results from this study will be reported at a later date.

The fifth local closure experiment compared the measured cloud condensation nuclei (CCN) supersaturation spectrum with the CCN supersaturation spectrum derived from Köhler theory applied to the measured aerosol number and chemical mass size distributions and TDMA growth factors. This intercomparison addressed questions of aerosol activation that affect the indirect radiative influence of aerosols on climate. The droplet-nucleating potential of the aerosol population was measured directly with a thermal diffusion cloud chamber that subjects the aerosol to a variety of supersaturations and counts the number of droplets that are nucleated. This defined the number concentration of CCN as a function of the supersaturation spectrum, CCN(S%). The supersaturation spectrum of the aerosol population was predicted independently using the aerosol number and inorganic and organic chemical mass size distribution, certain simplifying assumptions (e.g., an externally mixed aerosol and insoluble content), the aerosol growth characteristics determined from the TDMA (intercomparison 2), and Köhler theory. Results from Cape Grim indicate closure was achieved for marine air masses but that calculated CCN overpredicted the measured CCN in more continental air masses [Covert *et al.*, this issue].

Finally, the sixth local closure experiment compared the measured cloud droplet number concentration just above cloud base and the cloud droplet number concentration calculated from the measured updraft velocity, aerosol number size distribution, aerosol chemical composition, and CCN supersaturation below cloud base. This closure study added the complexity of real clouds

to the measurements of closure study 5 and tested the ability of cloud microphysical models to predict cloud droplet number concentrations from properties of the subcloud aerosol as well as the suitability of parameterizations of cloud droplet number concentration used in global-scale models. The methods used to determine the CCN supersaturation spectrum do not reproduce the time history of supersaturation encountered in real clouds. The cloud droplet size distribution just above cloud base (but high enough to be above the level of peak supersaturation) was calculated using the observed updraft velocity in two ways, from the measured CCN supersaturation spectrum and from the measured aerosol size distribution and chemical composition. The extent of agreement between the measured and calculated droplet number concentrations and size distributions provided a valuable indicator of the suitability of current models and measurement approaches for representing the initial stages of cloud droplet nucleation and growth [Boers *et al.*, this issue; Yum *et al.*, this issue].

A column closure experiment extends the local (zero-dimensional) closure to multiple altitudes (one-dimensional closure) in order to compare and calibrate satellite and surface-based column-integrated radiation measurements with in situ (aircraft) aerosol chemical, physical, and radiative measurements. Specifically, question 2, Can the measured physical and chemical properties of the aerosol in a vertical column be used to accurately predict the integrated effect of aerosols on radiative transfer?

A column closure experiment requires the integration of a large number of measurements [Quinn *et al.*, 1996; Russell *et al.*, 1997], and the ACE 1 column closure analysis is still in progress. The analysis is using the in situ measurements of particle scattering at several altitudes to compute the clear-sky column-integrated particle scattering. The airborne aerosol lidar is being used to scale the in situ measurements over appropriate altitude intervals and to identify any layering that the in situ measurements might have missed. The column aerosol scattering derived from in situ measurements will be compared with estimated aerosol optical depths from the AVHRR satellites and from ground and ship Sun photometers. The column closure experiments thus provide an important database with which to test and calibrate remotely sensed and indirectly determined aerosol properties for climate models.

Apart from the direct effect of aerosols on climate, aerosols in the lower atmosphere are known to control climate indirectly through their influence on the cloud droplet number concentration. The depth of the cloud, together with the cloud droplet number concentration, define cloud optical depth (essentially cloud column closure) which is a quantity closely related to cloud albedo. By observing aerosol below clouds, cloud droplets at several levels inside the clouds, and upwelling and downwelling short wave radiation above clouds, a unique data set is obtained linking subcloud aerosol to a climate pa-

rameter (cloud albedo) in the pristine environment of the Southern Ocean. Furthermore, cloud albedo is a parameter which can be retrieved from satellite observations. Thus the in situ data set can be used to test cloud optical depth retrieval algorithms.

Column closure experiments are also needed for reasons totally unrelated to climate. The impact of atmospheric aerosol scattering must be estimated and corrected for in order to derive information such as water-leaving radiances from satellite sensors. The atmospheric correction algorithms that have been developed for this purpose [Gordon and Wang, 1994] require assumptions about the nature and concentration of the aerosol and its resulting integrated radiative impact. The ACE 1 column closure experiments will help to improve the aerosol/radiative corrections. The transit flights between the United States and Australia will provide aerosol chemical and physical data to improve the regional aerosol corrections.

The area near Cape Grim, Tasmania, was chosen for ACE 1 in part because it provides an opportunity to characterize the natural aerosol and hence a background from which to quantify anthropogenic perturbations. However, measuring aerosol radiative effects in this background low-optical-depth case (0.04 to 0.05 [Forgan, 1990; Durkee *et al.*, 1991]) presents a significant challenge requiring extremely careful calibrations and intercalibrations. Current optical instruments (nephelometers and Sun photometers) are capable of making accurate and precise measurements in this region, but the signal-to-noise ratio is small.

To provide a contrasting natural high-optical-depth case to validate the column closure experimental approach, an additional experiment was conducted near Hawaii on the transit to Australia. This experiment used the Kilauea volcano plume, which provided a well-contained, soot-free source of sulfate aerosols with optical depths of 0.1 and higher. The degassing associated with Kilauea's ongoing eruptive phase (since 1983) constituted a point source for sulfur gases and associated sulfate aerosol into the relatively pristine atmosphere of the central Pacific. The plume provided a strong signal that was readily detected by satellite and in situ measurements. The radiative closure experiment carried out both during descent through the plume and in the adjacent clean air will provide a range of responses for the in situ instrumentation aboard the C-130, including the specific effect of markedly increased sulfate concentrations superimposed on a background aerosol. The direct comparison with adjacent clean air will help to quantify the importance of aerosol sulfate in aerosol extinction and will help to refine the algorithms needed to interpret satellite data by providing data at a much larger aerosol optical depth.

5.2. Process Studies

Objective 2 is to quantify the physical and chemical processes controlling the formation, evolution, and

fate of the major aerosol types and how these processes affect the number size distribution, the chemical composition, and the radiative and cloud nucleating properties of the particles. Three specific questions were addressed as part of ACE 1 in order to quantify the biological, chemical, and physical processes controlling the source, evolution, and fate of aerosol particles in the marine boundary layer. Question 3 is, What are the biological, chemical, and physical processes controlling the concentration of DMS in surface seawater and its flux to the atmosphere?

The background non-sea-salt sulfate aerosol over the oceans is thought to be largely derived from DMS [Charlson *et al.*, 1987]. To evaluate the importance of this natural aerosol source and to predict how this source may change with a changing climate, climate models require quantitative descriptions of the biological, chemical, and physical processes that are involved. At this time, the processes controlling DMS production and air-sea exchange are highly uncertain.

DMS is one component of an active seawater sulfur cycle [Bates *et al.*, 1994]. The precursor of DMS, DMSP (dimethylsulfonium propionate), is produced by many phytoplankton species as an osmolyte. During phytoplankton senescence and/or grazing, DMSP is released to the water column and enzymatically cleaved to DMS and other sulfur compounds. DMS can be lost from the water column by air-sea exchange, microbial consumption, or photochemical oxidation. The factors controlling the rate of DMS cycling in surface seawater and hence its seawater concentration strongly affect the amount of DMS that is released to the atmosphere. Although the seawater sulfur cycle component of ACE 1 was ultimately not funded, the biological, chemical, and physical oceanographic measurements made aboard *Southern Surveyor* as part of the JGOFS provided a regional descriptive picture of the oceanographic conditions during ACE 1 (F. B. Griffiths *et al.*, manuscript in preparation, 1998) and some insights into the processes that affect DMS concentrations [Jones *et al.*, this issue].

The air-sea flux of DMS is the starting point of the marine atmospheric sulfur cycle and thus limits the amount of sulfur available for aerosol formation and growth. The uncertainties associated with calculating oceanic DMS fluxes from seawater DMS concentration are roughly a factor of 2 [Yvon *et al.*, 1996] and thus impose a significant uncertainty in chemical transport models. To reduce these uncertainties, the flux of DMS to the atmosphere during ACE 1 was calculated by four independent methods: seawater DMS concentrations coupled with air-sea exchange models [Bates *et al.*, this issue], the atmospheric diurnal cycle of DMS concentrations coupled with atmospheric photochemical models [De Bruyn *et al.*, this issue], a micrometeorological flux/gradient method [Russell *et al.*, this issue], and a mixed layer budget approach as part of the Lagrangian experiments [Mari *et al.*, this issue; Russell *et al.*, this issue; Suhre *et al.*, this issue]. The results from the lat-

ter three methods are consistent with the lower end of the range calculated using air-sea exchange models.

Question 4 is, What are the rates and efficiencies of the processes controlling DMS and SO₂ oxidation in the marine boundary layer? The oxidation rates and conversion efficiencies of the various sulfur species are critical input parameters for calculating sulfate aerosol column burdens in aerosol/climate models [Langner and Rodhe, 1991; Erikson *et al.*, 1991; Benkovitz *et al.*, 1994; Taylor and Penner, 1994; Tarrason *et al.*, 1995; Pham *et al.*, 1995; Chin and Jacobs, 1996; Feichter *et al.*, 1996; Chuang *et al.*, 1997; Kasibhatla *et al.*, 1997]. While the oxidation of DMS is the presumed driving force for the production of submicron marine aerosols, the details of this process are still controversial. Although virtually all published models assume a high efficiency for conversion of DMS to SO₂ and nss sulfate, measurements in Kiribas suggest that most equatorial DMS is converted to SO₂ but less than half of this DMS actually becomes nss sulfate [Bandy *et al.*, 1996; Huebert *et al.*, 1996a]. Is SO₂ the principal product of DMS oxidation? Is any nss sulfate formed directly? What fraction of the SO₂ in the marine boundary layer is removed through deposition to the ocean surface and onto sea-salt particles, and what fraction is oxidized to sulfate aerosol [Sievering *et al.*, 1992]? Answers to these questions require field observations that will challenge mechanistic models.

Time series data can be very useful in elucidating sulfur chemistry, particularly if the air is sufficiently horizontally homogeneous that advection is not the primary cause of concentration changes. During ACE 1, both Cape Grim and *Discoverer* supported extensive measurements of sulfur chemistry that have been used to quantify processes controlling sulfur gas chemistry. DMS and SO₂ measurements aboard *Discoverer* suggest that DMS to SO₂ conversion efficiencies decrease with increasing latitude (W. J. De Bruyn *et al.*, manuscript in preparation, 1998). Photochemical box model calculations suggest the yield of SO₂ from DMS was 30–50% in the region south of Australia [De Bruyn *et al.*, this issue]. Aerosol chemical measurements at Cape Grim suggest that the supermicron non-sea-salt sulfate was formed predominantly by the ozone oxidation of SO₂ in sea-salt aerosol water (H. Sievering *et al.*, unpublished manuscript, 1998).

Lagrangian experiments offer tremendous potential for studying these atmospheric oxidation processes and chemical budgets. During the Atlantic Stratocumulus Transition Experiment (ASTEX), the Marine Aerosol and Gas Exchange (MAGE) Science Team tested a Lagrangian experimental approach for observing changes in the chemical species in the marine boundary layer near the Azores [Huebert *et al.*, 1996b]. Initial results suggested that this approach can yield process information without the confounding effects of air mass changes.

The Lagrangian strategy was developed further during ACE 1. Two different forecast-trajectory models were used to test possible Lagrangian start times and locations. To meet the ACE 1 objectives, the ship and aircraft needed to be able to reach the starting point, the air mass needed to avoid contact with land, and the air mass needed to stay within aircraft range for more than 1 day. Measurements aboard the ship were used to characterize the meteorological conditions and aerosol properties of the starting air mass. The Lagrangian experiment was initialized with the launch of "smart" balloons to mark air parcels for repeated study from the C-130. These constant-volume balloons contained an internal bladder into which air could be pumped or removed, in order to adjust the density and maintain a nearly constant altitude (S. Businger et al., manuscript in preparation, 1998). The balloons broadcasted their GPS-derived locations and several meteorological parameters every few minutes.

Two Lagrangian experiments were conducted. During Lagrangian A (Figure 13), one balloon was launched from the R/V *Discoverer* prior to sunrise. The C-130 then flew circular legs just to the north of this balloon across sunrise and again late in the afternoon. Although the aircraft was unable to pick up the signal from the balloon during a third flight, the flight legs were flown where the forecast trajectory had predicted the balloon would have been. To ensure that Lagrangian B (Figure 14) would begin in cloud-free air, the aircraft sought out a clear area and began flying patterns before the three balloons were launched from *Discoverer*. The aircraft was able to sample in the vicinity of the balloons for three flights (over 30 hours). The balloons maintained their relative positions in spite of a large change in wind speed and direction, suggesting that the air mass maintained its identity quite well. This conclusion also is supported by several sets of chemical and physical data. The concentrations of nss sulfate and methanesulfonate both increased throughout the day and remained constant or dropped slightly at night, as would be expected for photochemically de-

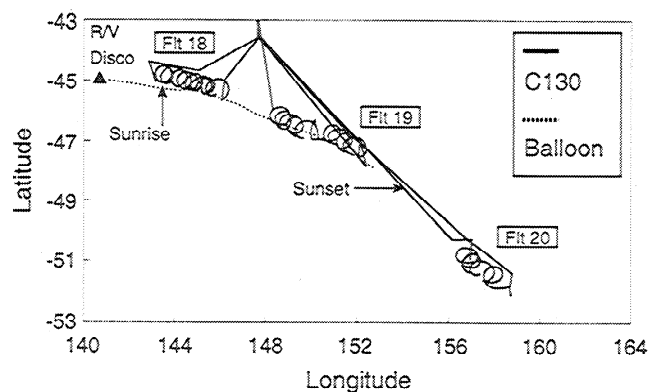


Figure 13. C-130 flights and balloon tracks for Lagrangian A.

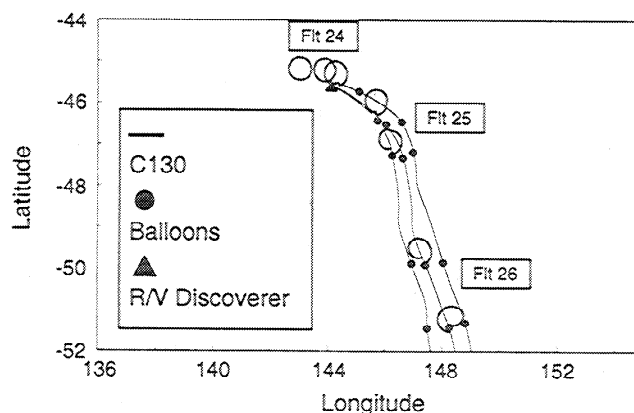


Figure 14. C-130 flights in the lower MBL and balloon tracks for Lagrangian B.

rived species. Changes in DMS and SO_2 concentrations also were consistent with photochemical processes. The planetary boundary layer during both Lagrangian experiments was divided into a surface mixed layer (referred to here as the boundary layer) of 500–800 m depth and a more stable layer (the buffer layer) above it that had many of the chemical characteristics of the boundary layer but mixed only slowly with the boundary layer and the free troposphere. Exchange between the layers was quantified in several ways, including an analysis based on the appearance in the boundary layer of an aerosol mode that previously had only existed in the buffer layer [Russell et al., this issue]. Although the thorough analysis of data from these Lagrangian experiments cannot be completed in time for this special section, it is already clear that by observing the evolution of boundary layer chemistry in an air mass throughout a diurnal cycle, the Lagrangian budget analysis will be able to constrain both surface fluxes and process rates.

Question 5 is, What are the rates and efficiencies of the processes controlling the nucleation, growth, distribution, and removal of particles in the remote marine atmosphere? The partitioning of DMS oxidation products between new particle production and particle growth will affect the submicron aerosol size distribution and, in turn, the effect of these particles on climate. Parameters that determine whether gaseous species condense to form new particles or condense onto existing particles include the saturation vapor pressure of the condensing species (H_2SO_4 , MSA, and NH_3), RH, temperature, and the existing particle number concentration and surface area [McMurry and Stolzenburg, 1989].

The marine boundary layer aerosol generally offers a large amount of surface area upon which gases such as H_2SO_4 can condense. It is therefore a rare event when the condensable vapors reach sufficiently high supersaturations to nucleate new particles in large concentrations [Covert et al., 1992; Hoppel et al., 1994]. Since precipitation, cloud processing, and coagulation are continually removing particle number, some source

of new particles is needed to explain why the total number remains relatively constant over time.

Observations of high ultrafine particle ($3 \text{ nm} < D_p < 20 \text{ nm}$) number concentrations in the clean upper tropospheric air and process-model calculations suggest that this region may be a source of new nuclei [Ito, 1985; Clarke, 1993; Raes *et al.*, 1993; Raes, 1995; Weber *et al.*, 1995; Weber and McMurry, 1996]. Air in cumulus updrafts may be scavenged of nearly all its aerosol mass, resulting in an air mass with low particle surface area. In addition, low temperatures at these altitudes reduce saturation vapor pressures, thus promoting bursts of nucleation, followed by coagulation. Subsidence and entrainment can then bring these fresh nuclei into the boundary layer.

During ACE 1, measurements of the number size distribution between 3 and 600 nm diameter were used to identify events of new particle production and mixing in both the marine boundary layer and free troposphere. Simultaneous measurements during ACE 1 of both precursor gases and ultrafine particle concentrations confirmed the photochemical formation of new particles in the outflow from cumulus clouds [Clarke *et al.*, this issue] that has been observed elsewhere [e.g., Radke and Hobbs, 1991; Perry and Hobbs, 1994]. New particles were observed to mix into the marine boundary layer after cold frontal passages and during periods of synoptic-scale subsidence and convective mixing between the free troposphere and the marine boundary layer [Bates *et al.*, this issue; Brechtel *et al.*, this issue]. Chemical mass size distributions, the ammonium to nss sulfate molar ratio as a function of size, and single particle analysis were used to determine the role of specific chemical species in new particle production and the conditions that inhibit particle production [Weber *et al.*, this issue].

In addition to homogeneous nucleation and subsequent growth through condensation, the aerosol size distribution can be transformed by cloud processing. Submicron aerosols can cycle through nonprecipitating clouds many times before being removed from the atmosphere through rain [Hoppel *et al.*, 1986]. During a cycle, the larger particles are activated and become cloud droplets. In addition, gas phase species and smaller particles can be absorbed into the droplets. When the cloud droplet evaporates, the residue is larger than the original particle. The result is a size distribution that contains two submicron peaks, one due to homogeneous nucleation and one due to cloud cycling. The thin layer of stratocumulus clouds that often existed over the ACE 1 study region provided an opportunity to investigate the effect of cloud cycling on the size distribution. These measurements will be used to estimate the fraction of sulfur species oxidized in cloud.

5.3. Modeling Activities

Objective 3 is to assess the climatic importance of remote marine aerosols. Question 6 is, How can observa-

tions be used to improve the accuracy of aerosol-climate models?

The climatic importance of the atmospheric aerosol requires improved parameterization of the processes that control the aerosol sources, properties, evolution, and spatial distribution. While satellites offer observations on a planetary scale, they lack any capability for chemical analysis of the aerosol particles and for coupling the spatial distribution of the aerosol to spatially and temporally variable aerosol precursors and source processes. Modeling of the aerosol system, including gaseous precursors and sources, aerosol production processes and properties, and the three-dimensional spatial and temporal distribution offers the only possibility for coupling locally measured properties to global forcing and effects.

The observational data collected during ACE 1 will be useful for testing and improving a hierarchy of related models [Penner *et al.*, 1994]: (1) detailed process models to describe DMS production and air-sea exchange, atmospheric gas and liquid phase chemical reaction rates, and aerosol and cloud microphysical processes and properties, (2) one-dimensional single-column models to evaluate the performance of process parameterizations and to provide information on the vertical profiles of aerosol parameters needed for radiative transfer calculations, (3) cloud scale three-dimensional large-eddy simulation (LES) models with explicit cloud microphysics to assess the interaction between clouds and aerosols, and (4) regional and global three-dimensional models to reveal the geographical and temporal distributions of the aerosol. Rather than simply providing the modeling community with data, ACE 1 was planned in concert with members of the modeling community (Table 2) to ensure that the data would be useful for validating and refining models.

6. Data Archive

The development and maintenance of a comprehensive and accurate data archive is a critical step in meeting the ACE goals. The overall ACE data management philosophy is to make the completed data set available to the world research community as soon as possible in order to better incorporate aerosols into global climate models. A centralized data archive has been established to combine the entire ACE 1 data set, including the measurements listed here (Table 1), the regional atmospheric soundings (Figure 2), satellite images, and the meteorological products generated for the experiment. This integrated database will allow users a single access to the variety of measured and derived fields obtained during ACE 1. The data archive can be accessed at <http://www.joss.ucar.edu/>.

7. Conclusions

Although the ultimate success of the experiment can only be fully gauged after a more complete data analysis

Table 2. Modeling Studies

Modeling Studies	Investigator	Institution
MBL sulfur/aerosol evolution	Frank Raes and Rita Van Dingenen	CEC Environment Institute, Ispra
MBL sulfur/aerosol evolution	Dean Hegg and Marcia Baker	University of Washington
MBL sulfur/aerosol evolution	Lynn Russell	Princeton University,
	Spyros Pandis, and	Carnegie Mellon University,
	John Seinfeld	California Institute of Technology
DMS production model	Albert Gabric and	Griffith University
	Francesca Campolongo	
UPRC water nucleation model	Boris Gorbunov, R. Hamilton, and	Middlesex University
	K. Sabelfeld	
Cumulus ensemble model	Steve Krueger and	University of Utah,
	Steve Siems	Monash University
LES stratocumulus cloud model	Yefim Kogan	Oklahoma University/CIMMS
MBL photochemistry model	Ron Prinn and Alex Pszenny	Massachusetts Institute of
		Technology
Aerosol chemical and optical properties	Patricia Quinn	NOAA/PMEL
Radiative transfer model	V. Ramaswamy	NOAA/GFDL
Satellite/aerosol model	Antony Clarke,	University of Hawaii
	John Porter, Barry Huebert, and	
	Philip Durkee	Naval Postgraduate School
Chemical transport model	Prasad Kasibhatla,	Duke University,
	Doug Davis, and William Chameides	Georgia Institute of Technology
Coupled chemical transport—climate model	Jost Lelieveld and Frank Dentener	University of Utrecht
Mesoscale Chemical Transport Model	Robert Rosset, Karsten Suhre, and	Laboratoire D'Aerologie
	Celine Mari	
Global chemical transport/aerosol model	Stephen Schwartz and	Brookhaven National Laboratory
	Carmen Benkovitz	

period, it is very clear from the initial results that most of the experiment's objectives were met. The data and analyses, reported in this special section, describe many of the results.

1. Measurements of the chemical, physical, radiative, and cloud nucleating properties of the aerosols were made using state of the art instrumentation in a wide range of environments [Bates *et al.*, this issue; Baumgardner and Clarke, this issue; Berg *et al.*, this issue; Boers *et al.*, this issue; Boers and Krummel, this issue; Brechtel *et al.*, this issue; Clarke *et al.*, this issue; Covert *et al.*, this issue; Hudson *et al.*, this issue; Huebert *et al.*, this issue; Kreidenweis *et al.*, this issue; Middlebrook *et al.*, this issue; Murphy *et al.*, this issue; Quinn *et al.*, this issue; Quinn and Coffman, this issue; Russell *et al.*, this issue; Weber *et al.*, this issue]. This included mainly the background marine atmosphere, but also volcanic plumes of Kilauea, Hawaii, and Mount Ruapehu, New Zealand, and some biomass burning and anthropogenic air masses from Australia. These data formed the basis for closure studies that assessed our ability to calculate radiative and cloud nucleating properties of the aerosol from its fundamental properties, the chemical and physical size distributions [Quinn and Coffman, this issue; Covert *et al.*, this issue]. This detailed data set is now available (<http://www.joss.ucar.edu/>) to further develop and test aerosol parameterizations in regional and global climate models.

2. Clear-sky column-integrated particle scattering experiments were carried out over a wide range of at-

mospheric optical depths by including not only background marine atmosphere near Tasmania but the volcanic plume of Kilauea. The integration of this large data set will require additional time and will be reported in future publications (A. D. Clarke *et al.*, manuscript in preparation, 1998).

3. Detailed measurements were made of the biological and chemical characteristics of the water masses south and west of Tasmania. During the month-long intensive campaign, we were able to observe the evolution of these water masses as conditions changed from winter to spring. These data have been used to assess the rate of DMS released from the ocean to the atmosphere [Bates *et al.*, this issue]. Initial results suggest biological productivity and DMS concentrations were highest where seasonal thermoclines were being established and where mesoscale mixing of waters of subtropical and subantarctic origin was occurring [Jones *et al.*, this issue].

4. One of the more exciting aspects of this type of experiment was the real-time observations that directly addressed current hypotheses, for example, where new particle production occurs in the atmosphere. Simultaneous measurements during ACE 1 of both the precursor gases (sulfur dioxide, sulfuric acid, and ammonia) and ultrafine particle concentrations clearly showed the photochemical formation of new particles in the outflow of cumulus clouds and in regions of high precursor gas concentrations [Clarke *et al.*, this issue]. New particles were observed to mix into the marine boundary layer af-

ter cold frontal passages and during periods of synoptic-scale subsidence and convective mixing between the free troposphere and the marine boundary layer [Bates *et al.*, this issue]. The ultrafine particles were regularly observed during these meteorological events on *Discoverer* [Bates *et al.*, this issue] and at Cape Grim and Macquarie Island [Brechtel *et al.*, this issue; Kreidenweis *et al.*, this issue].

5. The value of Lagrangian studies for deconvoluting the effects of dynamics and chemistry was clearly demonstrated during ACE 1 [Huebert *et al.*, this issue; Mari *et al.*, this issue; Mauldin *et al.*, this issue; Russell *et al.*, this issue; Suhre *et al.*, this issue]. While such an experiment in the "roaring forties" where wind and weather conditions are known for their severity must be considered high risk, the potential pay-off was large. With the aid of outstanding meteorological support from the Australian Bureau of Meteorology, two Lagrangian experiments were conducted during ACE 1. The data have been used to quantify the rates and efficiencies of sulfur gas oxidation (D. Thornton *et al.*, manuscript in preparation, 1998) and the evolution of the aerosol [Huebert *et al.*, this issue; Russell *et al.*, this issue] over the 2-day periods of these experiments. Additional shipboard measurements of DMS and SO₂ diurnal cycles were used to constrain oxidation rates and efficiencies [De Bruyn *et al.*, this issue].

ACE 1 (<http://saga.pmel.noaa.gov/ace1.html>) was the first in a series of IGAC aerosol experiments. The understanding gained in this experiment is now being used to study progressively more complex environments. The Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) focused on clear-column closure experiments off the east coast of the United States in July 1996 (<http://geo.arc.nasa.gov/sgg/tarfox/>). ACE 2 extended the process and closure studies to the eastern North Atlantic Ocean in June/July 1997 and focused on the anthropogenic aerosols from the European continent and mineral dust from the African continent (<http://www.ei.jrc.it/ace2/>). Planning is now underway for the next ACE that will focus on the region downwind of the rapidly increasing pollution sources in eastern Asia (<http://saga.pmel.noaa.gov/aceasia/>). We hope the papers presented in this special section will serve as a stimulus to join both in the process of interpreting the unique ACE 1 data set and in future ACE.

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T. S. Bates, Pacific Marine Environmental Laboratory, NOAA, 7600 Sand Point Way NE, Seattle, WA 98115. (e-mail: bates@pmel.noaa.gov)

P. A. Durkee, Department of Meteorology, Naval Postgraduate School, Monterey, CA 93943.

J. L. Gras, Division of Atmospheric Research, CSIRO, Mordialloc, Victoria 3195, Australia.

F. B. Griffiths, Division of Marine Research, CSIRO, Hobart, Tasmania 7000, Australia.

B. J. Huebert, Department of Oceanography, University of Hawaii, Honolulu, HI 96822.

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